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CHARCOAL PERFORMANCE UNDER SIMULATED ACCIDENT CONDITIONS.(U)
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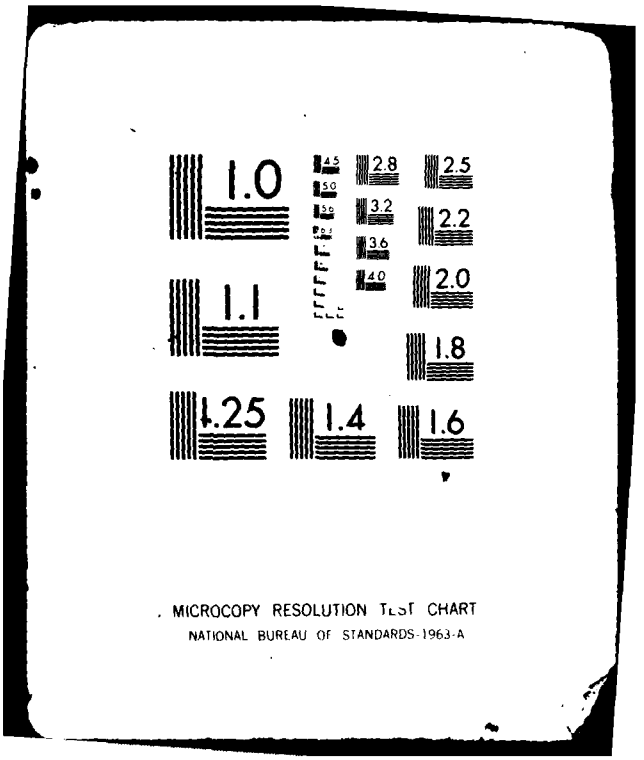
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Regulatory Guide 1.52 stipulates a radiation level for iodine buildup on the adsorber of 10^9 rads as one of the typical accident conditions for atmospheric clean up systems. The laboratory research in progress seeks to study the combined effects of in-service weathering, exposure to atmosphere contaminants, and radiation doses on the retention of iodine by the carbon. A number of service and weathered carbons have been exposed to the γ-radiation from the ^{60}Co source (approximately 1 MeV) and to the radiation

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20. ABSTRACT (Continued)

from the NRL LINAC facility (approximately 45 MeV). Total radiation levels of 10^7, 10^8 and 10^9 rads were used and the carbons were evaluated before and after irradiation by the methyliodide-131 penetration test (30°C, ASTM-3803-79). Surprising improvements were observed after static (i.e. no air flow) exposures of the carbons to the irradiation. Flow-through exposures during irradiation are now in progress using air or argon flows and with or without methyliodide-127.

CONTENTS

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Appendix 1, describing the NRL Linear Accelerator, was prepared by Ralph Tobin, now in Code 4740.

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CHARCOAL PERFORMANCE UNDER SIMULATED ACCIDENT CONDITIONS

1. INTRODUCTION

At any stage in the service life of an activated carbon adsorber that is used in nuclear air cleaning applications, the carbon must have the capability to retain the airborne radioiodine that may be liberated under accident conditions. During normal operations there is a slow and continuous degradation in the efficiency of a service carbon (1,2) and, when accident conditions are superimposed, additional problems can be expected. These include possible radiation damage to the physical structure of the activated carbon, the desorption of adsorbed materials, induced radioactivity after exposure to the accident radiation, and chemical changes in the adsorbed contaminants.

The small magnitude of radiation present in a non-accident mode does not appear to influence the carbon, and the efficiency of the carbon adsorber is controlled by the intake of atmospheric and/or local contaminants. The adsorption units of engineered-safety-feature (ESF) atmosphere cleanup systems must operate under postulated DBA (design basis accident) conditions. Regulatory Guide 1.52 (Revision 2, March 1978) (3) tabulates the DBA average radiation level for airborne radioactive materials

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at 10^6 rads/hr in the primary system and a dose from iodine buildup on the adsorber of 10^9 rads. In a secondary system, the corresponding values are 10^5 rads/hr and 10^9 rads.

In an accident mode, the concentration of ^{131}I can increase manyfold above background levels and above the periods of normal operation, thus increasing the rates of chemical reaction with both the base carbon and the impregnated chemicals. The reaction of the iodine with the carbon networks is thus enhanced and can lead directly to the formation of organic-^{131}I within the carbon bed. The ^{131}I trapped within the carbon networks can slowly form the more volatile methyliodide and "bleed" into the effluent air at a rate that will be dependent on the accumulated carbon weathering, the impregnation chemicals, and the magnitude of the radiation dose. Several accident scenarios have been postulated and the subsequent carbon behavior is being studied.

Facilities are available at NRL to expose nuclear carbons to the high radiation levels present during a DBA. The carbons will be evaluated before and after the exposures and with and without humidified gas flow through the carbons.

The following program of experimentation will be undertaken:

1. Charcoals will contain KI_x, TEDA, and a combination of KI + TEDA impregnations.
2. The carbons will receive a range of weathering and exposure to atmospheric contaminants.
3. The exposure to Gamma-radiation will be studied under no-flow (static) conditions and in air flows at different dose rates for periods of 100 hours and for 30 days; CH_3I

penetration tests will be conducted during and after exposure.

4. The carbons will undergo static exposures in the Linac Accelerator for integrated doses to 10^9 rads, 10^8 rads, 10^7 rads, and 10^6 rads and the results will be compared with those using cobalt-60 radiation.
5. Identification of iodine species in the effluent stream from a charcoal adsorber will be determined by the argon matrix trapping technique.
6. The iodine penetration of similar carbons exposed to different dose rates and to different radiation sources in equivalent integrated doses will be compared.
7. If dose rate effects are significant, additional work to quantify the effect of dose rate will be planned.
8. The effect of noble gases on the iodine retention of carbons in a radiation environment will be determined.

2. REVIEW OF PUBLISHED WORK

2.1 Savannah River Laboratory

Methyliodide radiolysis tests, first reported by Milham and Jones (4) and by Jones (20), were concerned with the effect of gamma radiation (approximately 2×10^7 rads/hr) on activated carbon (Type 416, unimpregnated) while a steam-air-methyliodide mixture was flowing through the test bed. Variables other than RH were held constant in these preliminary tests. The reduction in penetration in the radiation field was interpreted as a decomposition of methyliodide by gamma radiolysis, followed by a strong retention of the elemental iodine that is formed. The gamma field decreased the penetrations for flows of 0.91 sec. contact time (τ). There was little effect on penetration with flows of τ = 0.18 sec. These observations are summarized in Table 1.

Table 1: Results of Methyliodide Radiolysis on Type 416 Carbon at SRL (4).

Gamma Field (rads/hr)	τ (sec)	RH %	Penetration %
0	0.91	50	19.8
2×10^7	0.91	50	2.4
2×10^7	0.91	50	1.5
0	0.91	68	46.7
2×10^7	0.91	64	14.8
0	0.18	78	47.3
2×10^7	0.18	65	64.5

The assumption that radiolytic decomposition of methyliodide into non-penetrating forms of iodine, i.e. elemental iodine, as the only factor causing reduced penetration, was reexamined by Evans and Jones (7). It was suggested that:

> "The carbon may inhibit the radiolytic decomposition of methyl iodide. During most of the residence time in the gamma field, the methyl iodide must be temporarily adsorbed on the surface of the test carbon. If a methyl iodide molecule is decomposed, the ions formed are not in the free gaseous state where they are likely to collide and react with other ions or molecules; they are in a partially immobilized adsorbed state where they are most likely to recombine because of their proximity. Thus, decomposition is inhibited."

The observed decomposition was compared with the published data of Tang and Castleman (21) who irradiated methyliodide-air mixtures of initial concentrations ranging from 1.1×10^{-8} to 2.4×10^{-4} molar and at dose rates of 2.77×10^{4} rads/hr to 1.06×10^{6} rads/hr. According to Tang and Castleman's results, 50% decomposition of the methyliodide should occur at 1.7×10^{7} rads/hr in about 25 seconds. At 27% RH and 23 sec residence time, 50% decomposition did take place. At 50% RH and 220 sec residence time, 35% decomposition was observed, but >99% was predicted from Tang and Castleman's results.

When elemental iodine was used on carbon in a radiation field of 1.5×10^{7} rads/hr and exposure times to 100 hours, Evans and Jones (9) found that the radiation dose rate influenced the rate of iodine desorption. An analysis was made of the iodine activity trapped on unimpregnated carbon back-up beds. They were able to collect about 20% of the activity in liquid nitrogen cold traps and analyses were made by gas chromatography. Four organic iodides were identified: methyliodide, methylene iodide, ethyl iodide and vinyl iodide. There were many other unidentified compounds. These results are the first recorded evidence of the "scrambling" of the iodine isotope into the molecular species

which are desorbed from activated carbon.

The new irradiation facility provided 4.7×10^7 rads/hr in the center of the empty tube and 3.3×10^7 rads/hr in the center of the carbon bed (10). Because of the increase in dose rate, it was anticipated that the radiolytic desorption in the new apparatus would be two to four times greater. The expected increase did not occur (11), which suggested a possible ionization saturation phenomenon at higher dose rates.

Two impregnated coconut-shell carbons (A with 5% TEDA and B with 2% TEDA and 2% KI) were subjected to a radiation desorption test in which the absorbed dose rate was 3×10^7 rads/hr. As in all these experiments, irradiation and the testing were done simultaneously. The elemental iodine loading was 1 hr., the purge 4 hrs., at 90-95% RH and 80°C. The results are summarized in Table 2 (12). After six months service, both carbons performed equally and better than non-impregnated carbon.

Table 2: Elemental iodine penetration with simultaneous irradiation (3×10^7 rads per hour) and testing (12).

Carbon Service (months)	Test Duration (hours)	Total rads	Penetration A	Penetration B
			%	%
0	5	1.5×10^8	.028	.034
6 (composite)	5	1.5×10^8	.048	.052
6 (inlet face)	5	1.5×10^8	.073	.090
0	27	8.1×10^8	.050	.051
6	89	27.0×10^8	.36	.36

Additional testing (13) with special carbons having different proportions of TEDA and KI showed that a 10x16 mesh coconut

shell activated carbon containing 1% TEDA, 2% KI, and a flame retardant was preferred by the investigators.

The SRL Radiolytic Desorption Test is: 1 hr loading time followed by 4 hr desorption time at 80°C and 95% relative humidity in a radiation field of ~2.7×10^7 rad/hr (adsorbed dose rate in carbon). Total I_2 loading of ~0.7 mg/g. Note that this is a simultaneous exposure of the carbon to the radiation field and to the test gas. In the SRL Screening Test Results (10), a number of unused carbons were evaluated for elemental iodine penetration by three different tests. Test 1 was the Radiolytic Desorption Test, Test 2 was a control test identical with Test 1 except without exposure to radiation, and Test 3 was made at a high temperature (180°C) without exposure to radiation. Some of the results are summarized in Table 3 (10). With two exceptions, the high temperature test was most severe in yielding penetration. The two exceptions, a base carbon and a TEDA + KI impregnated carbon were more vulnerable to Test 1. Test 2 as a control test at 80°C without radiation gave the least penetration and when compared to Test 3 made at 180°C demonstrates the enhanced chemical reactivity of the impregnant and the base carbon at the high temperature.

Radiolysis tests on Type GX-176 carbon, service-aged, (17) indicate the role of contaminants accumulated in service. This may not necessarily be equal to time in service due to seasonal variation in atmospheric contaminants. The high temperature test date (no radiolysis at 180°C) shows greater penetration than with radiolysis at 80°C. A more complete explanation depends on the

influence of contaminants on the rates of formation of the organic iodides.

Table 3: Penetration of elemental iodine evaluated by three tests (10).

	Penetration %		
	Test 1 (Radiation)	Test 2 (Control)	Test 3 (High Temp.)
BC 416 Coconut	.283	<.001	.004
BC 727 KI_3	.017	<.001	.052
G 617 KI_3	.014	<.001	.028
G 620 KI_3	.029	<.001	.056
Witco 42 KI_3	.058	.011	6.484
G 615 TEDA + KI	.037	<.001	.006
G 618 TEDA	.003	<.001	.003
Witco 19 TEDA	.008	-	12.03
Witco 36 TEDA	.014	-	4.56
MSA 85851 KI_3	.030	.001	18.08
Calsicat DI-131 KI_3	.045	-	2.412
GX 126	.021	<.001	.070
Dose time	1 hr	1 hr	10 min.
Temperature °C	80	80	23
Purge time	5 hr	4 hr	4 hr
Temperature °C	80	80	180

Additional comparisons can be made with the results for service-aged GX-176 carbon (18). A number of carbons were removed from confinement compartments where the carbons were aged for specified times. Table 4 presents some of these results for which the iodine penetration in radiolysis test data (80°C) are compared with the iodine retention at 180°C. At first, radiolysis at 80°C showed greater penetration than thermal desorption at 180°C, but with service-aging longer than about 18 months, the reverse situation was true. Again, the contaminants accumulated in the longer exposure times must enter into the

impregnant-carbon interactions. Evans (18) was able to establish a very positive correlation between the pH of the water exhaust and the thermal desorption at 180°C.

Table 4. I_2 penetration (%) for service-aged GX-176 (18).

	Aging (months)	Radiolysis** % (80°)	No Radiation* % (180°C)
Control	0	0.028	0.002
P-2	7	.062	.011
P-2	9	.050	.016
P-2	14	.121	.060
P-2	19	.228	2.30
P-2	30	.543	5.04
K-2	6	.052	0.027
K-2	12	.060	.093
K-2	18	.090	.409
K-2	24	.176	1.60
K-2	30	.334	6.35

* Elemental iodine penetration in 4 hrs at 180°C.

** Elemental iodine, 1 hr loading and 4 hr desorption at 80°C and 95% RH in radiation field of 1.5×10^7 rad/hr.

Evans (19) reported a valuable correlation between the methyliodide penetration (80°C) and radiolytic-iodine penetration (80°C and 1.5×10^7 rad/hr on the carbon). The results in Table 5 (19) show a steady increase in both penetrations as a function of service age. The methyl iodide penetration is always greater than the radiolytic elemental iodine penetration. The mean ratio was 330 with a standard deviation of 120 for carbons of service age up to 30 months, the maximum time in the SRP confinement system.

Table 5: Radiolytic penetration as a function of methyliodide penetration (19).

Service Age	Radiolytic* iodine penetration	Methyliodide penetration (80°C)	Ratio
months	%	%	
0	0.028	6.7	240
0	.028	6.4	230
6	.052	24.1	460
6	.052	24.6	470
7	.062	32.6	530
14	.121	52.9	440
19	.228	74.6	330
25	.270	89.7	330
27	.359	92.2	260
27	.414	92.9	230
30	.577	92.3	160
		mean	330
		std. deviation	120

* Iodine desorbed in 5 hrs at 80°C, 95% RH, and radiation dose $>1.5 \times 10^7$ rad/hr. in the carbon.

In conclusion, the SRL studies show the following facts in regard to the radiolysis of carbons:

1. Radiolytic desorption of elemental iodine forms several organic iodide species.

2. Below 10^7 rads/hr, the reaction is dose-dependent, and above this value, radiolytic desorption does not increase with increasing radiation intensity.

3. The methyliodide penetration (80°C) is several hundredfold greater than the penetration of elemental iodine at 80°C and 1.5×10^7 rads/hr due to radiolysis.

4. The gamma-induced decomposition of the methyliodide

molecule with the formation of iodine is not the only factor causing reduced penetration upon radiolysis.

5. The increase in dose rate from 1.7×10^7 to 3.3×10^7 rads/hr measured in the center of the carbon bed did not increase the expected radiolysis. It was suggested that a possible ionization saturation phenomenon might be present.

6. There is a steady increase of both methyliodide penetration (80°C) and radiolytic elemental iodine penetration as a function of service age and decreasing pH of the water extracts.

One important difference in the SRL studies and those at NRL is that in the former case, irradiation and testing were done simultaneously. At NRL the carbons were irradiated first and then tested with methyliodide-131.

2.2 Chemical Systems Laboratory (22)

Samples of whetlerite, a coal base-activated carbon impregnated with ammoniacal chromium and copper salts, were exposed to radiation doses of 0.5 to 100×10^6 rads. The test gas was cyanogen chloride (CK).

The following general observations were made regarding the results:

a. For the "base charcoal," twelve out of a total of 18 samples gave a specific adsorptivity greater than the controls. Of these twelve "improved" samples, seven exceeded the upper standard deviation level of the controls.

b. The dose range giving the most consistent improvement in the base charcoal samples was approximately 1×10^6 to 20×10^6 rads.

c. For the whetlerized charcoal, four out of the five samples tested showed a reduction in efficiency.

The author concluded (22): "Unfortunately, because of the many variables involved, including handling of the sample before and after irradiation, and in the testing procedure, the results showed a great deal of scatter." "In summary, we believe that this effort, while inconclusive in its immediate results, may have surfaced most of the problems that must be solved during future research in this area."

3. RADIATION EXPOSURES OF CARBONS TO THE γ-SOURCE ^{60}Co

The investigation is relevant to the post-accident period in which the radiation source would be the radioactive gases passing through the carbon adsorber. It is not possible to expose the carbons in the laboratory to a radioactive gas at the high Curie level of an accident. However, the carbons can be exposed to equivalent radiation levels (γ and/or β sources) and at the same time subject the carbon to gas flows of non-radioactive species in order to study the chemistry of the resultant degradation reactions.

The position for samples around the Cobalt-60 A-source at NRL is shown in Figure 1. The static exposures of the carbons to date have made use of the positions A-1, A-3, A-5, and A-7. The A-1 position is located within the cylindrical array of eight Co-60 rods and the remaining positions are outside of this array, as shown. The half-life of Co-60 is 5.2 years and the activity was essentially constant over the duration of each exposure. The calibrated intensities of the three positions in Figure 1 are:

A-1	7.5×10^5 rads per hour
A-2 to A-7	1.4×10^4 rads per hour
A-8 to A-19	2.5×10^3 rads per hour

The sample container is of stainless steel, 3 in. (7.62 cm) i.d., and the useful height in the radiation field was ±2.5 in., (±6.35 cm), measured from the center. This permits a sample volume of 580 cm^3. The closure at the top is made with a new

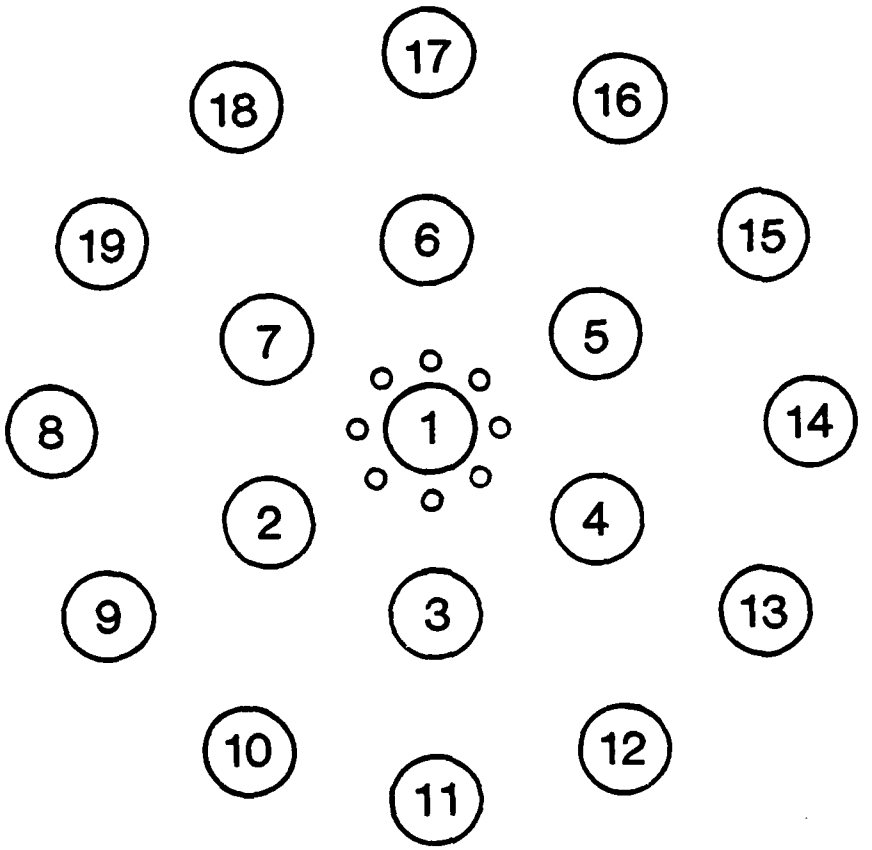

Figure 1: Position Array for Samples Around the Cobalt-60 A-Source at NRL. The A-1 position is located within the cylindrical array of eight Cobalt-60 rods as shown above.

3 in. (7.62 cm) O-ring (synthetic rubber) and the assembly lowered through a depth of 12 ft. (3.66 meters) water (22°C) to the desired location.

New Carbons

In earlier studies (1,2) on the weathering of impregnated carbons, it was found that the moisture retained by new, unused carbons after exposure had a degrading influence on subsequent tests for methyliodide-131 penetration. It was considered worthwhile, therefore, to irradiate several new carbons for about 100 hrs, with and without the added moisture and without air flow, in the γ-field of the Cobalt-60 and to determine the subsequent penetration of methyliodide-131.

The results are given in Table 6. Tests 1, 2, and 3 were made with dried carbons in the period 20-24 November 1981. The carbons, prehumidified in an air flow of 90% relative humidity (21-24 November), were then irradiated in tests 4, 5, and 6 for the period 24-28 November. The total radiation flux was approximately the same in both cases, namely 1.4×10^7 rads. In the subsequent penetration test with methyliodide-131, the same procedure was used for all samples. The air flow at 95% RH was maintained for two hours; the insult of methyliodide-131 in 95% RH was then added for two hours. Finally, the air purge at 95% RH was conducted for four hours.

The radiation damage to the above new carbons, as measured by methyliodide penetration, was not significant in that the penetrations were below the permissible level (ASTM Standard D4069-81). In two of the three carbons, prehumidification before radiation gave the same behavior as previously observed for prehumidified and non-prehumidified penetration tests.

Table 6: Penetrations of methyliodide-131 through three new carbons after exposure to γ-radiation from A-source of Cobalt-60.

Position in ^{60}Co Source		Carbon*	NRL Test	Exposure Time	Total Rads	Penetration %
1	A-3	615	5217	98 hrs	1.46×10^7	0.27±.02
2	A-5	727	5218	98 hrs	1.39×10^7	0.30±.11
3	A-7	S and S	5219	98 hrs	1.49×10^7	0.08±.06
		Prehumidified Carbon				
4	A-3	615	5221	96 hrs	1.38×10^7	0.10±.08 0.16±.08
5	A-5	727	5222	96 hrs	1.32×10^7	1.40±.84
6	A-7	S and S	5223	96 hrs	1.42×10^7	1.51±.84
		13 Jan. - 6 March (1981)				
7	A-1	G-615 new, as received	5228	52.15 days	9.03×10^8	0.07±.05

* See Appendix 2 for characteristics of these carbons.

A total exposure of 10^9 rads was completed with a sample of G-615 (KI and TEDA impregnation) placed in the A-1 position (see Table 6). The required time was estimated to be 56 days. The exposure was interrupted in short intervals for which an accurate log was maintained. The integrated intensity for the 52.15

days in the A-1 position (7.22×10^5 rads/hr, av.) was 9.03×10^8 rads. The penetration of methyliodide-131 after the long exposure was 0.07±.05%. This value is of the same order of magnitude previously observed for G-615 with radiation (see Table 6) and without radiation. It was concluded at this point in time that a new carbon that passes the ASTM qualification test is not subject to significant degradation under the conditions of the radiation exposure.

Exposure of Used Carbons

Since all carbons accumulate a considerable amount of contaminants during the time in service, the ionization of some of these contaminants in the radiation field could influence the retention of iodine compounds by activated carbons. Two used carbons were first irradiated, each at 10^7 and 10^8 rads, in the Co-60 facility. The penetration of methyliodide-131 was determined before and after the irradiation. The results are given in Table 7.

The NRL sample 5143 was a discard carbon removed from a utility operation where it had been in place for four years. The moisture content was 19.7% and the pH of the water extract was 7.3. It was installed as a KI_x-impregnated coconut carbon.

The NRL sample 5233 had been used for two years in a glove box operation in which iodine-125 was used to prepare organic preparations for medical treatments. The pH of water extract was 6.9 and there was no significant change in pH after exposure

to Co-60 radiation. The material when new was NACAR G 615 having a co-impregnation of TEDA (triethylenediamine) and KI (potassium iodide) on coconut shell carbon.

Table 7: Penetration before and after ^{60}Co irradiation.

NRL Sample	% Penetration of Methyliodide-131*		
	Original	10^7 rads	10^8 rads
5143	31.0	16.4	6.31
5233	30.1	11.7	7.25

* Standard Test ASTM/ANSI D3803-79

The exposure of the above two samples to the γ-radiation of ^{60}Co decreased the penetration of methyliodide-131. In other words, the efficiency for the removal of methyliodide-131 was increased. This is a remarkable observation and will be further supplemented by the results obtained after exposures of the same carbons to the LINAC radiation.

4. LINAC IRRADIATION OF CARBON SAMPLES AT NRL

Since the times required to reach 10^9 rads in the available positions of the ^{60}Co facility proved to be excessively long, the use of the NRL LINAC facility was explored with the cooperation of Dr. K. M. Murray (Code 6654). A description and the operating characteristics of the NRL LINAC are given in Appendix 1. The aluminum sample container used for the activated carbon exposures was cylindrical, 11 cm. diameter and 11 cm. high. The total volume of approximately one liter was divided into four equal sections (to accommodate four different samples) by inserting thin aluminum partitions having a snug fit with the inside diameter and with the inside height. Each section held about 210 cc of carbon. The container was mounted on a rotating base operating at about 5 rpm.

In order to determine the dose rate, three short tests (see Figure 2) were made by Dr. Murray and the LINAC staff on 7 January 1981. In the first two tests, an array of nine TLD's was placed at first 50 cm and then at 75 cm for about three seconds at each location. The 75 cm location resulted in a more uniform dose distribution than did the 50 cm and was chosen as the final distance. The dose count distribution as a function of distance is shown in Table 8 and Figure 4.

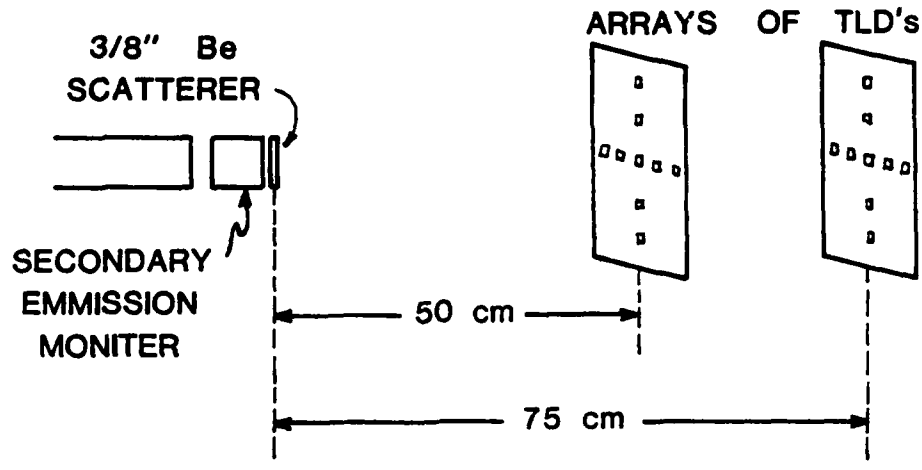

Figure 2: Alignment for determination of the dose rate.

Table 8: Dose Count of an Array of Nine TLD on Container

	50 cm	75 cm
TOP OUTER	1281	779
TOP INNER	3408	1427
RIGHT OUTER	2124	931
RIGHT INNER	2776	1458
BOTTOM OUTER	1987	793
BOTTOM INNER	3441	1119
LEFT OUTER	1503	700
LEFT INNER	2878	1249
CENTER	-	1573

To calibrate the beam monitor, a secondary emission monitor, SEM (see Figure 3), was used and three pairs of TLD's were placed in the sample which was rotated at 5 RPM. The sample was placed 75 cm from the scatterer and irradiated for ten seconds. The average current from the SEM for this period was 20 μA. This was integrated as shown below using a Keithley Model

610 CR electrometer, a voltage-to-frequency converter (VFC) and a scaler. With the VFC set at 100 HZ/V, the total count was 362.

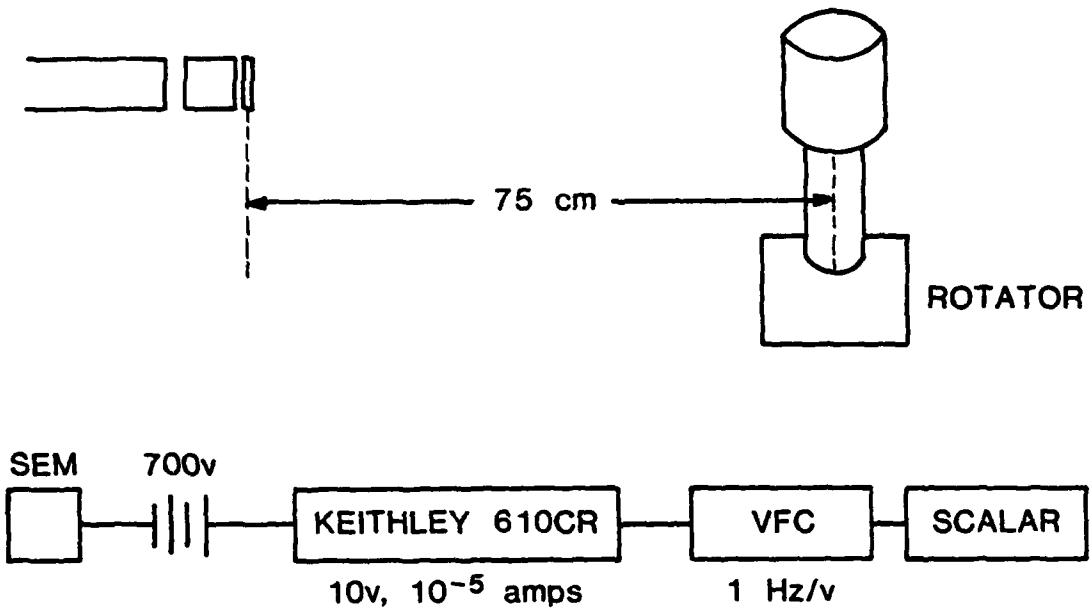

Figure 3: Calibration of Beam with a Secondary Emission Monitor SEM.

	Dose on Each TLD		
	# 1	#2	Avg.
CENTER TLD	27.0 Krads	28.0 Krads	27.5 Krads
MIDDLE TLD	17 Krads	20 Krads	18.5 Krads
TOP TLD	11 Krads	11 Krads	11 Krads

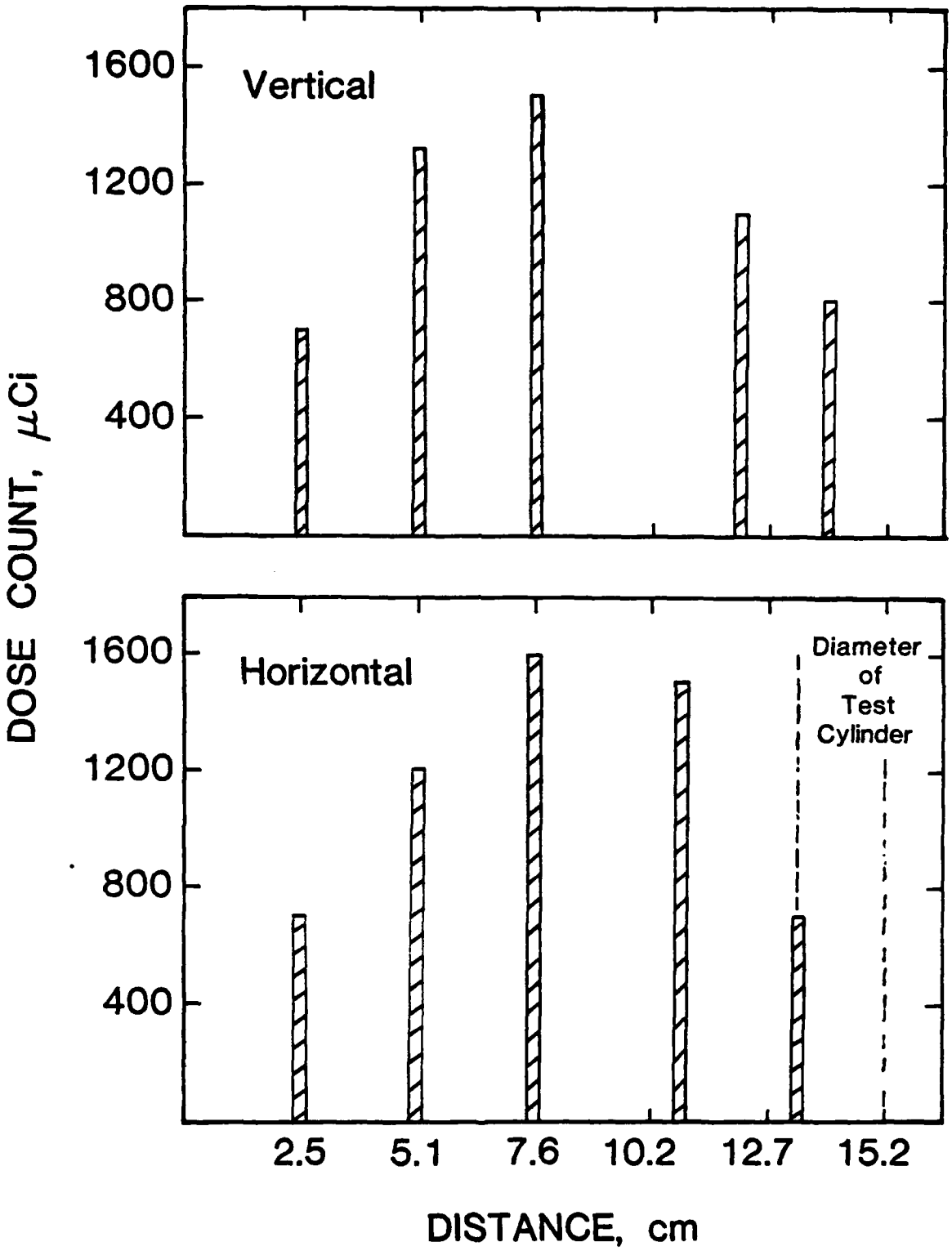

Figure 4: Distribution of Dose Count (μCi) as a Function of Horizontal and Vertical Distance at 75 cm from window.

Choosing a dose of 20×10^3 rads as representative of most of the sample, the value of 5.5×10^3 rads per count was obtained with the VFC set at 1 HZ/VOLT. The following counts correspond to the designated total rads that are desired:

1.8×10^5 counts	10^9 rads
1.8×10^4 counts	10^8 rads
1.8×10^3 counts	10^7 rads
1.8×10^2 counts	10^6 rads

Induced Activity after Irradiation

The first samples were irradiated continuously at NRL on 10-11 August 1981, resulting in 191,000 counts on the beam monitor. This corresponded to a total dose of 1.05×10^9 rads.

The irradiated samples were found to be radioactive. The presence of this activity raised a problem of interference in subsequent penetration tests with iodine-131 when crystal counting is used. Dr. K. M. Murray took approximately 1g from each of the four samples and made an analysis for the induced activity. The spectrum of each sample was found to be dominated by the gamma rays from I-126, as shown below:

1. KI + TEDA (G-615)	0.25μCi/g	50.9μCi/sample
2. KI + TEDA (G-615)	0.25μCi/g	46.7μCi/sample
3. KI + TEDA (BC-787)	0.27μCi/g	43.1μCi/sample
4. KI + TEDA (KITEG II)	0.14μCi/g	23.4μCi/sample

Linac exposures have also been made at 10^8 and 10^7 rads. Table 9 summarizes the results for the formation of radioiodine-126 which decreased about tenfold for each tenfold decrease in total rads of exposure.

The iodine-126 activity was attributed to a γ,n nuclear reaction with ^{127}I. In addition to I-126, other minor reactions were detected: I-127 (γ, 3n), I-124 and C 12 (γ, n) C11. The half-life of the above radioactive species are:

I-126 13.2d C-11 20.5m

I-124 4.2d

Table 9: Formation of Iodine-126 in Linac Exposures.

Irradiation		Measured		Calculated to t = 0
Date	NRL	μC_I	Date	μC_I
10-11 Aug 81 at 10^9 rads	5242	21.71	27 Aug	50.9
	5243	20.99	26 Aug	46.7
	5244	18.39	27 Aug	43.1
	5245	10.52	26 Aug	23.4
20 Oct 81 at 10^8 rads	5251	2.02	3 Nov	4.26
	5252	1.73	5 Nov	4.06
	5253	1.99	2 Nov	3.98
	5254	2.00	3 Nov	4.22
20 Oct 81 at 10^7 rads	5255	0.174	12 Nov	0.593
	5256	0.314	2 Nov	0.628
	5257	0.166	12 Nov	0.566
	5258	0.242	5 Nov	0.567

The initial intensity of iodine-126 A(o) was calculated from the measured value A(t) at time t as follows: $A(t) = A(o)e^{-kt} = A(o)e^{-0.693\ t/t_{1/2}}$, where $t_{1/2}$ is the half-life. Sufficient iodine-126 activity was found to invalidate a penetration measurement based on a sodium iodide crystal counting procedure. This is due to the interference of the Compton Scattering from ^{126}I into the channel used for the ^{131}I measurement.

Gamma-ray analyses were carried out utilizing 10% efficient Ge-Li detectors and a Canberra Series 80 multichannel analyzer system used in conjunction with a PDP 11/34 computer (Nuclear Environmental Services, 3 Choke Cherry Road, Rockville, MD 20850).

Exposure of Used Carbons

A number of used carbons from service were exposed on the LINAC facility to an accumulated value of 10^9 rads (32 hours). The penetrations of methyliodide-131 were determined using the Ge-Li detector and the results are given in Table 10.

Table 10: Penetration of Methyliodide-131 before and after LINAC exposure of 10^9 rads (10-11 Aug 1981)

NRL Sample	Weight (g)	Penetration, %		Ratio
		Initial	Radiated	
5233	160	30.1	0.039	1000
5234	147	10.5	0.033	320
5214	143	9.99	0.03	330
5151	141	38.9	0.002	1900

The NRL sample 5233 has been described above. NRL sample 5234 had been used for one year in a glove-box operation in which I-125 was used to prepare organic-iodine preparations for medical treatments. The original material was NACAR-G615, having a co-impregnation of TEDA (triethylenediamine) and KI (potassium iodide) on coconut shell carbon.

NRL sample 5214 had been weathered at NRL by passing outdoor air (1.05×10^6 cubic feet) from 17 Sept 1980 to 25 June 1981 (0.77 year). The pH of the inlet layer was 4.2 and that for the exit layer was 9.5. The original material was BC 787 which had

been impregnated with TEDA and KI on coconut shell carbon.

NRL sample 5151 had been weathered at NRL by passing outdoor air (2.79×10^6 cubic feet) from 30 May 1979 to 25 June 1981 (2.07 years). The pH of the inlet layer was 2.1 and that for the exit layer was 8.0. The original material was designated as Nucon KITEG II, Lot 024.

The recovery of carbon efficiency for trapping methyliodide-131 appears to increase with the quantity of contaminants added to the carbon adsorber during service. The ratio of the penetration of the initial carbon to that observed after irradiation is plotted in Figure 5 as a function of time in service. There is a definite trend relating the improvement of the carbon with the service time.

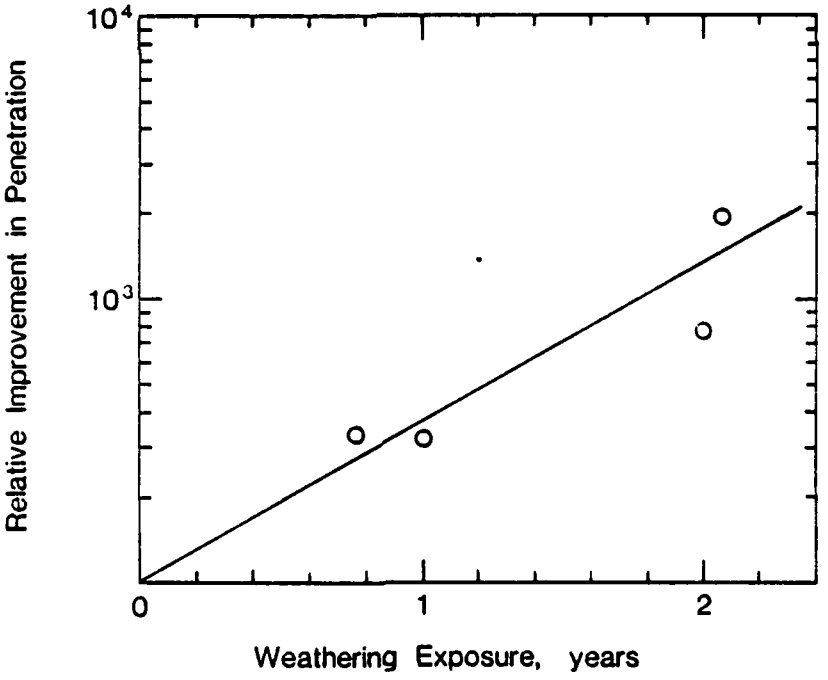

Figure 5: Ratio of the Penetration of the Initial Carbon to that observed after Irradiation as a Function of Time in Service.

During the methyliodide-131 penetration measurements (ASTM D-3803, 79), it was found that some of the induced I-126 was detected in the two back-up beds. The amounts (see Table 11) are small, but it is evidence of the presence of isotope "scrambling" among the species ^{126}I, ^{127}I, and ^{131}I present on the carbons during the test procedure. The mechanism is quite complex and involves both gaseous and adsorbed species.

Table 11: Migration of Induced ^{126}I during the Test Procedure (ASTM D-3803) for Trapping Methyliodide-131 at 30°C.

NRL Sample No.	Post-test Count for ^{126}I (μC_i)			Counting Date
	Test Sample	Back-up A	Back-up B	
5233	16.13	8.89(-3)	3.29(-5)	31 Aug
5234	18.14	1.98(-3)	<5.6(-5)	31 Aug
5214	13.45	4.89(-3)	2.54(-5)	1 Sept
5251	9.38	2.48(-3)	2.67(-5)	28 Aug

Measurements made by Nuclear Environmental Services, 3 Choke Cherry Road, Rockville, Md 20850

5. DISCUSSION AND CONCLUDING REMARKS

The exposure times in the NRL studies vary widely with the available radiation sources. In the Co-60 exposures, there are two positions:

Position	Intensity	Desired Exposures	
A-1	7.5×10^5 rads/hr	rads	time
		10^9	55 days
		10^8	5.6
		10^7	0.56
		10^6	0.056
A-2	1.4×10^4 rads/hr	10^9	2980
		10^8	298
		10^7	29.8
		10^6	2.98

With the present geometry of LINAC operations, which yields 3.1×10^7 rads/hr, the following exposures are required:

Desired Exposures	
Rads	Time
10^9	32 hrs
10^8	3.2 hrs
10^7	0.32 hrs
10^6	0.032 hrs

Figure 6 shows the wide variation in the duration of the exposures. To get a reasonable exposure time for a total of 10^6 rads on the LINAC, it will be necessary to increase the distance between the source and the sample target (presently 75 cm). It

would then be possible to get equal exposure times with both facilities and also to get a dose rate in between the Cobalt-60 and the present LINAC geometry. Regulatory Guide 1.52 (3) specifies a dose rate of 10^6 rads/hr in the primary locations and 10^5 rads/hr in the secondary locations.

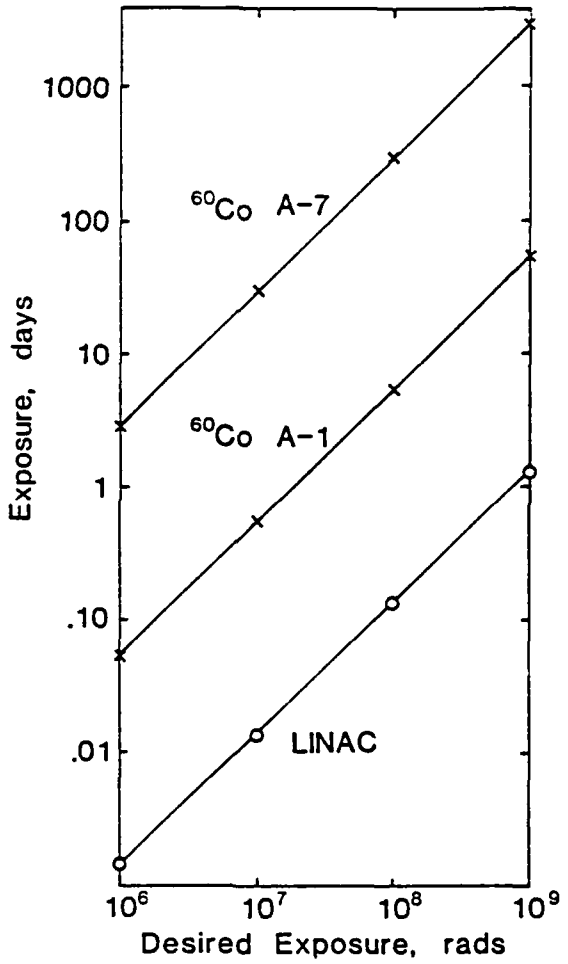

Figure 6 : Variation of Exposure with Different Sources as a Function of Desired Exposure.

The static irradiations of fifteen carbons have been completed at 10^7, 10^8, and 10^9 rads. Of these 45 samples, methyl-iodide-131 penetration values remain to be determined for 16 samples.

The exposures of carbons to humidified air flows with the gamma-radiation of Cobalt-60 are in progress. These are time-consuming experiments with exposure times up to 100 hours. The combined influences of continuous humidified air flows and the gamma radiation field of Cobalt-60 on service carbons are being varied. Total radiation levels of 10^6, 10^7, and 10^8 rads were used. Changes in methyliodide-127 penetration are followed daily by monitoring with gas chromatography using the electron capture detector for a dose period (2 hrs) and a purge period (4 hrs). The penetration of methyliodide increases with the duration of the humidified air flow (23) and decreases with the time of exposure to the radiation. The observed results are interpreted in terms of the degree of balance of these two opposing tendencies.

It will be possible to conduct tests to determine the effect of non-radioactive argon and/or krypton in contact with the carbon during irradiation. The ionization of the noble gas may interact with the contaminants of a used carbon, and influence the subsequent measurement of penetration using methyliodide-131.

It has been shown that KI_x and KI + TEDA impregnations may generate I-126 on exposure to radiation of 10^7 rads and above. It has yet to be demonstrated how TEDA impregnations alone will behave.

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APPENDIX 1

NRL LINEAR ACCELERATOR
U.S. Naval Research Laboratory
Washington, DC 20375

CHARACTERISTICS

The Naval Research Laboratory (NRL) Linac is a three-section, S-band accelerator which operates in the energy range 5 to 65 MeV. At maximum efficiency the beam energy is 42 MeV and peak beam current is 0.5A. Pulse width can be varied from 0.03 to 1.4 μs.

TEST PARAMETERS

Linac parameters and operating charateristics are typically very flexible. Data presented here are representative of normal opertions. The experimenter can arrange with the NRL Linac supervisor for operation of the Linac in other than routine modes.

Operating Characteristics

Operating characteristics of the NRL Linac are given in Table 12.

TABLE 12. OPERATING CHARACTERISTICS

Microwave Frequency	2853 MHz
Number of Accelerating Sections	3
Energy Range	5 to 65 MeV
Maximum Efficiency Operation:	
Beam Energy	42 MeV
Peak Beam Current	0.5 A
Rated Average Power	8 kW
Beam Pulse Width (FWHM)	0.03-1.4 μs
Pulse Repetition Rates per Second (single pulses are available within width range)	5, 15, 30, 60, 130, 360
Average Beam Current	
Port 1	250 μA
Port 2	100 μA (max)
Port 3	100 μA
Port 4	100 μA
Energy Spread, $\Delta E/E$[(a)]	0.4 to 10%
Transient Mode Operation:	
Beam Energy (ave)	30 MeV
Peak Beam Current	1 ampere
Beam Pulse Width (FWHM)	50 ns

(a) Value sensitive to beam energy and current.

Electron-Beam Loading

Electron-beam loading characteristics are given in Figure 7.

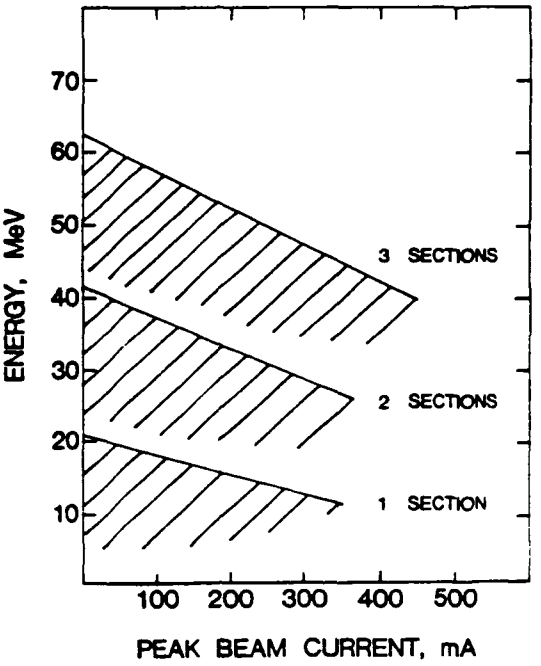

Fig. 7 — Beam loading characteristics

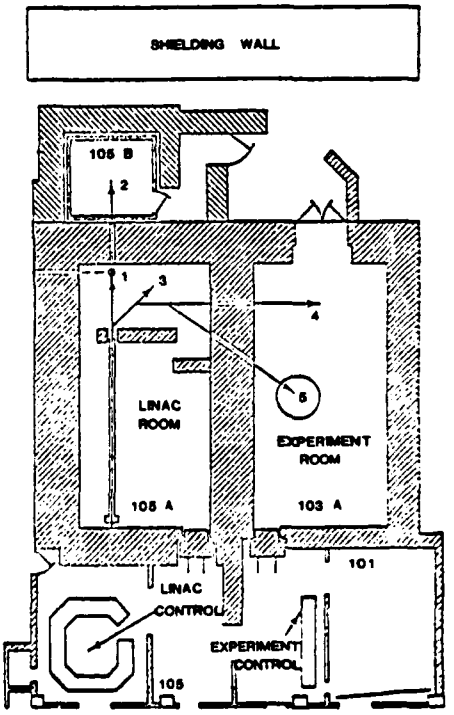

Fig. 8 — Floor plan of NRL LINAC laboratory

As with all Linacs, the electron current available over the entire energy range is not constant, being maximum near the center of the range and approaching zero at the upper limit. In the case of the NRL Linac, energies below 5 MeV are nearly impossible to obtain, and at 5 to 7 MeV unanalyzed beam currents are limited to 100-200 mA, peak. Due to the fact that the energy spread of the raw beam may be as large as 15 to 20 percent (FWHM), deflection and collimation of the beam reduces the available current.

Pulse Shape

The pulse shape for longer pulses (0.5 - 1.4 μs) in beam one are approximately rectangular. As the pulse length is reduced the shape is more triangular. Pulses appearing in beams 3 and 4 are somewhat less regular in shape due to the energy-time relationship within the duration of the pulse. Best rise time achieved on short pulses is 14 ns, although this could be improved with sufficient demand.

Repetition Rate and Pulse Predictability

Repetition rates are given in Table 12. Single pulses can be delivered for any pulse width within the normal pulse width range.

Pulse reproducibility and predictability are excellent. For example, on single pulse exposures, pulse amplitude predictability of ± 5 percent is typical with dark current background radiation less than 0.5 percent for a single 4×10^4 rads (Si) pulse.

Electron-Beam Geometry and Energy Spread

The size and shape of the beam spot depend on the energy, the current, the particular beam port employed, and the amount of focusing used. Spot diameter is typically in the range 0.2 to 1.0 cm. The shape of the spot can be made round or elliptical through the use of quadrupole focusing lenses. Extra large spot sizes can be produced by scattering from a foil.

The experimenter should be aware that radiation dose values quoted as characterizing the environment output are directly related to beam spot size; higher values are associated with smaller beam diameters.

Beam energy width E/E (FWHM) of the raw beam is 10 percent at 38 MeV and 1.0 percent at 56 MeV for the equilibrium mode. The energy width of deflected beams is adjustable by means of slits in the magnetic analysis system. The transient mode featuring high peak currents of short duration would exhibit a FWHM of 20 percent to 30 percent.

Neutron Irradiations (Requires technical assistance; call for details)

A compact target of water-cooled tantalum plates is available for neutron irradiations. To emphasize fast neutrons, a minimum of water is used. Three to four inches of lead is employed to reduce gamma contamination. Preliminary spectral measurements indicate a broad peak at about 1 MeV, with a differential flux of about 7×10^{12} neutrons/cm^2 per coulomb of Linac beam at a point 90° to the beam direction, 11 cm from the center of the target. Gamma contamination at this point is about 4×10^4 rads (Si) per coulomb of Linac

beam. A coulomb of 42 MeV electrons is collected in about 6 hours. (See Reference (3)).

Environment

The NRL Linac electron beam can be used directly as a radiation source, converted to X-rays by means of a bremsstrahlung target, or converted to neutrons by means of a photoneutron target.

Output Characteristics

The electron beam emerges through a water-cooled stainless steel window and can be used directly or converted to X-rays and neutrons. By appropraite experimental techniques either of these three components can be emphasized and their effect on various media studied.

TABLE 13 ENVIRONMENT CHARACTERISTICS

Electrons	
Peak current (50 ns pulse)	1 amp
Peak current (1.4 μs pulse)	0.5 amp
X-Rays (10 cm from converter plate [a])	1×10^3 rads (si)/pulse
Neutrons	1×10^6 1 MeV equivalent neutron/cm²/pulse

Measurement Techniques and Errors

Electron beam current measurements are made in two ways. One of these consists of a current transformer through which the electron beam passes just before emerging from the water-cooled exit window. The second involves Faraday cup techniques employed after the beam has emerged through the window. Both give results having errors less than ± 5 percent. (See Reference (4)).

Absorbed dose measurements are made using thermoluminescent detectors and reverse biased silicon diodes. These are calibrated against an absolute silicon calorimeter (5) or a ^{60}Co source.

Neutron intensity measurements are available using sulfur disks that give neutrons per cm² for energies above 3 MeV. Neutron spectrum information is also available. Dose measurements for neutrons are made using Li^6 and Li^7 enriched TLD's (Li^6F_2, Li^7F_2).

For the dose measurements involving use of the reverse biased silicon diode, digital readout is available giving total integrated dose for each pulse.

(a) Measured 11.5 cm from center of target and behind 4-inch lead gamma ray shield.

Electrical Noise

The electrical noise within the Linac laboratory space can be separated into two general categories:

1. Noise not synchronous with Linac radiation pulse, and
2. Noise synchronous with Linac radiation pulse.

Each of these categories can be subdivided into

(a) Radiated noise, and
(b) Conducted noise.

Nonsynchronous Noise

The most significant source for noise in this category is that associated with the silicon controlled rectifiers (SCR's) used in the waveguide cooling system for the Linac. The conducted component appears as several ''gliches'' of about 4 volts amplitude and 50 s duration appearing on the 115 VAC service lines.

The typical radiated component is such that a signal having a maximum amplitude of 3 millivolts peak-to-peak, a characteristic frequency of 4 kHz and lasting for 5 milliseconds is induced in a 60-foot length of semi-flexible, 1/2 inch-diameter coax cable with the cable terminated at its input end with 50 ohms.

Synchronous Noise

The most predominant source of noise synchronous with the Linac radiation pulse is that associated with the klystron modulators. The conducted component appears on the 115 VAC service lines with a maximum amplitude of 5 volts peak to peak with an average frequency of about 1.5 MHz and lasting for 20 μs.

The radiated component (measured within Room 101) will induce a signal in an inductive loop 5 inches in diameter and terminated in 50 ohms having a maximum amplitude of 15 millivolts with a frequency of 14 MHz and lasting for 10 μs.

By various means periodic improvements in all of these noise levels have been made and will continue to be made.

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APPENDIX 2

Impregnated Nuclear Carbons under Investigation

Nuclear Carbon	Nominal Size	CCl_4 Activity*	Source	Impregnation
BC 787	8x16	60	Coconut	Iodine salts and tertiary amines
BC 727	8x16	90	Coconut	KI + I_2
BC 717	8x16	60	Coconut	KI + I_2
G 615	8x16	60	Coconut	KI + TEDA
G 617	8x16	95	Coconut	KI + I_2
NSA (463563	8x16	60	Coconut	KI + I_2
AAF 2701	8x16	60	Coconut	KI + I_2
KITEG	8x16	60	Coal	Iodine Salts and tertiary amines
KITEG II, NUSORB	8x16	60	Coconut	
Sutcliffe, Speakman & Co.	8x16	60	Coal	5% TEDA

* ASTM D3467; TEDA = triethylenediamine.

APPENDIX 3

List of Figures

APPENDIX 4

List of Tables

NRC FORM 335 (11-81) U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER (Assigned by DDC): NUREG/CR/2550

4. TITLE AND SUBTITLE (Add Volume No., if appropriate): CHARCOAL PERFORMANCE UNDER SIMULATED ACCIDENT CONDITIONS

2. (Leave blank)

3. RECIPIENT'S ACCESSION NO.

7. AUTHOR(S): Victor R. Deitz

5. DATE REPORT COMPLETED: MONTH June, YEAR 1982

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code): Naval Research Laboratory, Washington, DC 20375

DATE REPORT ISSUED: MONTH March, YEAR 1982

6. (Leave blank)

8. (Leave blank)

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)

10. PROJECT/TASK/WORK UNIT NO.: 61-1427-0-1

11. FIN NO.: 87232

13. TYPE OF REPORT: Interim report on a continuing NRL problem.

PERIOD COVERED (Inclusive dates)

15. SUPPLEMENTARY NOTES

14. (Leave blank)

16. ABSTRACT (200 words or less)

Regulatory Guide 1.52 stipulates a radiation level for iodine buildup on the adsorber of 10^9 rads as one of the typical accident conditions for atmospheric clean up systems. The laboratory research in progress seeks to study the combined effects of in-service weathering, exposure to atmosphere contaminants, and radiation doses on the retention of iodine by the carbon. A number of service and weathered carbons have been exposed to the γ-radiation from the ^{60}Co source (approximately 1 MeV) and to the radiation from the NRL LINAC facility (approximately 45 MeV). Total radiation levels of 10^7, 10^8 and 10^9 rads were used and the carbons were evaluated before and after irradiation by the methyliodide-131 penetration test (30° C, ASTM-3803-79). Surprising improvements were observed after static (i.e. no air flow) exposures of the carbons to the irradiation. Flow-through exposures during irradiation are now in progress using air or argon flows and with or without methyliodide-127.

17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS

Charcoal
Removal of radioactive iodine
Irradiation
Methyliodide testing

17b. IDENTIFIERS/OPEN-ENDED TERMS

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